

A beta-alpha coincidence counting system for measurement of trace quantities of ^{238}U and ^{232}Th in aqueous samples at the Sudbury Neutrino Observatory

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A beta-alpha coincidence counting system for measurement of trace quantities of ^{238}U and ^{232}Th in aqueous samples at the Sudbury Neutrino Observatory.

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ABSTRACT: The Sudbury Neutrino Observatory experiment was built to measure the total flux of ^8B solar neutrinos via the neutral current disintegration deuterium nuclei. This process can be mimicked by daughter isotopes of ^{232}Th and ^{238}U which can photodisintegrate the deuterium nucleus. Measurement of the concentration of such radioisotopes in the heavy water was critical to the success of the experiment. A radium assay technique using Hydrous Titanium Oxide coated filters was developed for this purpose and it was used in conjunction with a delayed beta-alpha coincidence counting system. The design, calibration and operation of this counting system are described in this paper. The counting efficiency for ^{232}Th (^{224}Ra) and ^{238}U (^{226}Ra) were measured to be $50 \pm 5\%$ and $62 \pm 7\%$

KEYWORDS: Scintillators, scintillation and light emission processes (solid, gas and liquid scintillators); Very low-energy charged particle detectors

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1 Introduction

The Sudbury Neutrino Observatory (SNO) experiment was a heavy water Cherenkov detector built to investigate the discrepancy between the predicted and measured flux of ^8B solar neutrinos. Located approximately 2100 m underground (5890 m water equivalent) in the Vale Creighton Mine, Sudbury, Ontario, Canada, it consisted of a 12 m diameter acrylic vessel which held 1,000 tonnes of heavy water (D_2O). The acrylic vessel was surrounded by 7,000 tonnes of ultra-pure water (H_2O) shielding and approximately 9,500 photomultiplier tubes (PMTs) were used to detect Cherenkov light produced by neutrino interactions. Solar neutrinos interacted in the D_2O via one of three distinct processes: Charged Current (CC) $\nu_e + d \rightarrow p + p + e^-$, Neutral Current (NC) $\nu_x + d \rightarrow p + n + \nu_x$ or Elastic Scattering (ES) $\nu_x + e^- \rightarrow \nu_x + e^-$. The CC process is sensitive to electron-type neutrinos only, the type produced by the Sun, but the NC process is equally sensitive to all light active flavours. A non-unity ratio of CC flux to NC flux would be a direct indication of flavour changing neutrino oscillations. A detailed description of the SNO detector is given in [1].

To ensure a high signal-to-background ratio, ultra-pure water was used to minimize backgrounds from naturally occurring radioisotopes. Both the ^{232}Th and ^{238}U decay chains contain daughter isotopes (^{208}Tl and ^{214}Bi respectively) that emit gamma-rays with energies greater than 2.2 MeV. Such gammas can photodisintegrate a deuterium nucleus, producing a neutron which is indistinguishable from one produced by an NC neutrino interaction. Levels of radioactive contamination in the water were required to be such that less than one neutron per day was produced via photodisintegration. This was equivalent to approximately 10% of the neutrons produced per day from neutrino NC reactions.

Concentrations of ^{208}Tl and ^{214}Bi were measured in two independent ways: using data collected whilst the detector was measuring neutrinos (*in-situ*) and chemical assay of water extracted from

the detector (*ex-situ*). Three separate *ex-situ* methods were developed to measure ^{224}Ra (pre-cursor of ^{208}Tl) and ^{226}Ra (pre-cursor of ^{214}Bi) and ^{222}Rn , a possible cause of disequilibrium in the ^{238}U chain between ^{226}Ra and ^{214}Bi . To assay Ra, two independent methods were developed: MnOx [2] and HTiO [3, 4]. Due to leaching of trace contaminants in detector materials, Ra and Rn were not in equilibrium with their long-lived parent isotopes. To facilitate comparison between different measurement techniques, concentrations of Th and U were calculated assuming that equilibrium existed in the chain. The limits of one neutron produced per day via photodisintegration were equivalent to concentrations of $3.8 \times 10^{-15} \text{ g}^{232}\text{Th}/\text{gD}_2\text{O}$ and $3.0 \times 10^{-14} \text{ g}^{238}\text{U}/\text{gD}_2\text{O}$ in the heavy water region. For the light water shielding between the photomultiplier support structure and the acrylic vessel, purity requirements were less stringent as the probability of a gamma from a radioactive decay in this region photodisintegrating a deuterium nucleus was far smaller. Concentration requirements for the light water region were $3.7 \times 10^{-14} \text{ g}^{232}\text{Th}/\text{gH}_2\text{O}$ and $4.5 \times 10^{-13} \text{ g}^{238}\text{U}/\text{gH}_2\text{O}$.

To measure concentrations of ^{224}Ra and ^{226}Ra in the water, Hydrous Titanium Oxide (HTiO) was deposited onto a pair of filters housed in custom made polypropylene housings. These were connected to the underground water systems, and water from the detector was passed through them for a pre-determined period of time. During this time, heavy ions present in the water were extracted by the filters. Following extraction, the filters were taken to the surface laboratory where Ra isotopes were removed from them by flowing 15 litres of 0.1 M hydrochloric acid through them. The resulting acidic solution was concentrated to a volume of a few millilitres and added to 42 g of liquid scintillator. The concentration of radioactivity in the sample was measured using a custom-made beta-alpha coincidence counting system [5]. The original system was constructed from Nuclear Instrument Modules (NIM) and Computer Automated Measurement and Control (CAMAC) components. Due to its age, it was becoming increasingly prone to failure. A new system, constructed from custom designed printed circuit boards was manufactured by the Physics Electronics Workshop at the University of Oxford. In addition to being reliable, this “new” system was easy to use and portable. This paper will describe the principles behind the counting technique, the system and results from assay measurements during the SNO experiment.

2 Beta-alpha coincidences

The beta-alpha system was designed to measure the number of beta-alpha coincidences arising from the decay of daughter nuclei in the ^{232}Th and ^{238}U chains. Toward the end of the ^{238}U chain, ^{214}Bi decays by beta emission to ^{214}Po which subsequently alpha decays with a half-life of $164 \mu\text{s}$. The signature of this coincidence is a beta decay with a Q -value of 3.27 MeV followed many μs later by a 7.7 MeV alpha ($Q = 7.83 \text{ MeV}$). A similar coincidence exists in the ^{232}Th chain, where ^{212}Bi decays with a 64% branch by beta emission to a short lived state of ^{212}Po which alpha decays to ^{208}Pb with a $0.3 \mu\text{s}$ half life. The signature for this decay is a beta decay with a Q -value of 2.25 MeV followed a fraction of a μs later by an 8.78 MeV ($Q = 8.95 \text{ MeV}$) alpha. The difference between the half-lives of the Po isotopes can be used to separate between the ^{238}U and ^{232}Th decay chains.

To reduce the rate of accidental coincidences, good discrimination between alpha and beta events is required. Differences in the decay time of the liquid scintillator can be used to distinguish alpha and beta events in the scintillator. For organic scintillators, the timing profile of each event

can be described by several exponential decays arising from the singlet and triplet excited states. Singlet state de-excitation occurs quickly and is associated with the fastest timing component of the scintillator. Triplet states cannot decay directly to the ground state and a number of de-excitation channels are available. For example, two triplet state molecules could collide resulting in occupation of a singlet state which can decay directly to the ground state. The triplet de-excitation processes occur over a longer time period and are therefore associated with the longer timing components. The proportion of each timing component depends upon the ionization density of the incident particle. Due to an alpha particle's high ionization density and resultant ionization quenching, a high proportion of triplet relative to singlet states are produced, so the proportion of fast light is reduced relative to slow light for alpha particles. Beta particles are much less ionizing, resulting in a much higher proportion of fast light compared with slow.

A larger fraction of the total charge deposited by the pulse will be present in the tail of alpha events when compared to beta events. Discrimination is possible by taking the ratio of the charge observed in the tail of the pulse to that observed in the whole pulse. The difference in pulse shape can be used to discriminate between alphas and betas, which reduces accidental coincidences from sources such as beta-beta, beta-gamma and beta-noise coincidences. This technique is commonly referred to as pulse shape discrimination and it was used as the basis for this beta-alpha counting system, an overview of which is given in the following section.

3 System overview

The beta-alpha coincidence counting system can be broken down into three subsystems, namely PMT, beta-alpha electronic unit and the data acquisition PC. A liquid sample (~50 ml) was held in a small polymethylpentene jar¹ which was optically coupled to the surface of a 5 cm diameter photomultiplier tube.² Scintillation light produced by decay of radioisotopes in the sample was detected by the PMT.

Each PMT was housed in a 2.5 cm thick shield made from Oxygen Free High Conductivity (OFHC) copper, to reduce accidental coincidences arising from soft room gammas, lined with reflective foil. The housed PMTs were held in a high voltage cage which provided grounding for the shields and a high voltage interlock which prevented the PMTs from being opened when the high voltage was on. A maximum of eight PMTs could be used with the system.

The PMT output was connected to a custom-made beta-alpha board via a filter box. The filter box consisted of a pi network attenuator in conjunction with a 330 pF capacitor to smooth the pulse and in the process the pulse amplitude was attenuated by about a factor of two. This ensured pulses rose and fell smoothly, which was necessary for an accurate determination of the fraction of charge in the tail of the pulses. The output of the filter box was connected to a beta-alpha board which performed a series of logic operations on each event to determine whether or not a genuine beta-alpha coincidence had occurred. If a coincidence was detected, information from the board was read out and stored by the DAQ PC. In total eight boards were held in a crate, which provided power, and read out via the backplane. A schematic diagram of the system is shown in figure 1.

¹The jars were machined on a lathe to ensure they were flat.

²Electron tubes Ltd. 9266KB.

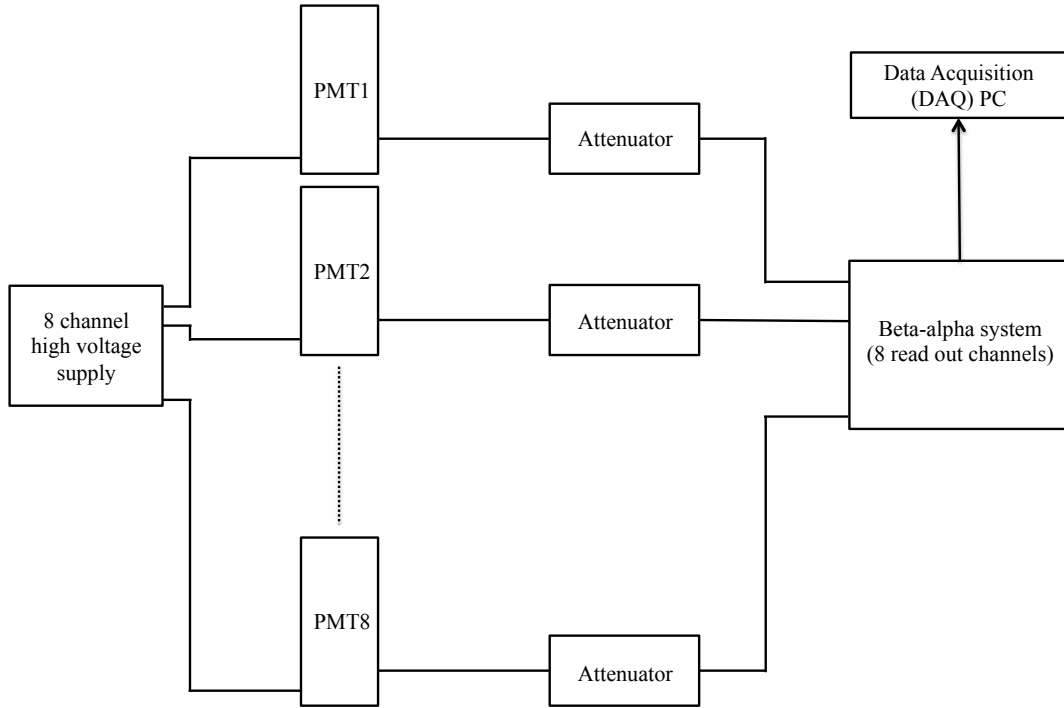


Figure 1. Schematic of the PMT and beta-alpha system.

3.1 Pulse handling of the boards

Any signal received by the board was subjected to a number of pre-determined threshold, timing and logic constraints to determine whether or not it was part of a beta-alpha coincidence. Logic operations were controlled by a Field Programmable Gate Array (FPGA) chip. The FPGA governed the communication between the boards and DAQ PC and contained a 40 MHz clock which was the reference point for all timings associated with the board. There were three thresholds and three timing intervals associated with a board. These are now discussed in turn, beginning with the thresholds.

RUN. The amplitude of every (smoothed) PMT signal entering the board is compared to this threshold. Only those pulses with amplitudes above this threshold were accepted and stored by the electronics. This threshold helped to reduce the number of noise pulses entering the system, however a small fraction of genuine beta pulses was lost, resulting in a lower system efficiency. The exact setting of this threshold was a compromise between noise rejection and beta acceptance.

ALPHA. Only the second pulse in the coincidence was subjected to this threshold. As the energy spectrum of the alpha pulses did not extend to zero (due to the two body nature of the decay), a carefully chosen non-zero threshold setting would not significantly affect the acceptance of genuine alpha pulses. The threshold was set such that nearly all of the alpha energy spectrum was accepted and low energy noise rejected. If the second pulse did not pass this threshold, the system was reset. The alpha threshold was optimised for the $0.3 \mu\text{s}$ Bi-Po coincidence from the ^{232}Th chain. As

the alpha from the ^{238}U chain Bi-Po coincidence is less energetic, this lower the acceptance of U chain events.

MU. This is an upper limit on pulse amplitude, designed to remove large pulses from the data. Such signals are usually produced by cosmic muons interacting in the scintillator, hence the name of the threshold. Any coincidence where either pulse crossed this threshold was discarded and the electronics reset.

If the RUN threshold was crossed by the incoming signal from the PMT, a voltage ramp and integrator were started. The amplitude of the voltage ramp decreased linearly as a function of time. By comparing the amplitude of this voltage ramp to three set thresholds, three timing intervals for charge integration were defined relative to the start of the voltage ramp. These integration times were called FIRST, SHORT and LONG and are now discussed.

FIRST. The FIRST threshold defined the time interval over which the first (beta) pulse was integrated. It extended beyond the peak of the pulse to the point where the amplitude had fallen to approximately 50% of its maximum value. In this system, this was set to 31 ns. This timing interval was set to ensure that the system deadtime between the first (beta) and second (alpha) pulses was as small as possible and that the resolution and shape of the beta spectrum were not affected significantly. When the voltage ramp crossed the FIRST threshold, signifying the end of the integration time period, the charge stored on the integrator was read out. The system then waited for a second pulse to pass the RUN threshold. The time between pulses was measured by the number of clock cycles that elapsed between the two pulses that triggered the RUN thresholds. The clock cycle time was 25 ns and the average minimum time between a beta and an alpha pulse in a beta-alpha coincidence that could be detected was roughly 65 ns.

SHORT. The SHORT threshold was set such that all but the tail of the second (alpha) pulse was integrated. This corresponded to integrating the first 50% of the pulse or equivalently 80% of the total pulse charge. The SHORT threshold was set such that the integration time was 49 ns.

LONG. The LONG threshold defined the time interval over which the full second (alpha) pulse was integrated. It was set to extend just beyond the full tail of the alpha pulse. The fraction of charge in the tail of the pulse, f_t was calculated offline using the following formula

$$f_t = \frac{Q_{\text{Long}} - Q_{\text{Short}}}{Q_{\text{Long}}}$$

where Q_{Long} is the integrated charge of the entire second pulse and Q_{Short} is the integrated charge of the first 49 ns of the second event.

The thresholds and timing intervals of each board were set using a double pulser which provided fake coincidences. The amplitude of the first pulse was set to approximately 10 mV, the width of each pulse was about 60 ns and the time interval between pulses was 100 ns. All thresholds and timings were adjusted using the appropriate potentiometer on the board. The thresholds and timings are now discussed in the context of an example coincidence.

3.2 An example coincidence

An example coincidence and associated timing graphs are given in figure 2. In this example, it is assumed that both pulses are above the RUN and ALPHA thresholds, but below the MU threshold. Therefore, both pulses will be detected by the board.

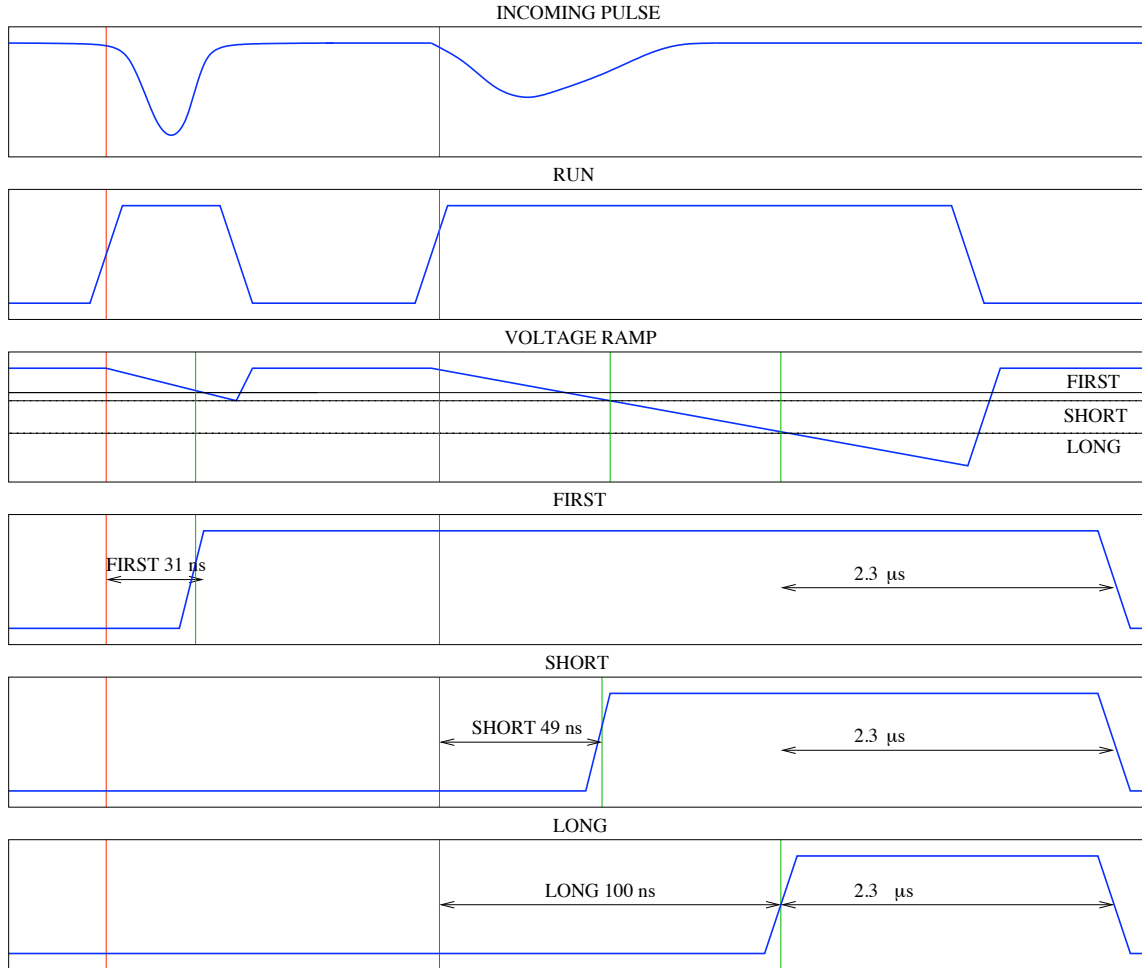


Figure 2. An example coincidence, with pulses separated by 100 ns. From top to bottom the illustrations are: the coincidence signal, the RUN threshold, the voltage ramp used for timing (with FIRST, SHORT and LONG thresholds marked), FIRST threshold, SHORT threshold and LONG threshold. For the FIRST, SHORT and LONG thresholds, the change in gradient is the point at which the voltage ramp crosses the threshold. The vertical red lines indicate when the integrator starts integrating. The time taken for the system to reset is 2.3 μ s as indicated at the end of the graphs.

The crossing of the RUN threshold by the first (beta) pulse starts a voltage ramp and integrator. This can be seen in figure 2 as the start of the beta pulse coincides with the switching of RUN to a positive value and the beginning of the voltage ramp. The voltage ramp decreases until it crosses the FIRST threshold, approximately 31 ns after the start of the beta pulse, causing FIRST to go positive. At the end of the beta pulse, RUN falls back to zero, the value of the integrator is stored on a capacitor and then the integrator is reset along with the voltage ramp. FIRST is not reset and remains positive. The system waits for another pulse.

If another pulse arrives within 1.6 ms a coincidence has been detected,³ RUN goes positive and the ramp and integrator are started again. In figure 2 a second pulse is detected. Approximately 49 ns after the start of the second (alpha) pulse, the SHORT threshold is crossed. The value of the integrator is stored on another capacitor, but the ramp and integrator are NOT reset. About 51 ns later (≈ 100 ns after the start of the α pulse) the LONG threshold is crossed, making SHORT positive. The value of the integrator is read out onto a third capacitor. All components are reset following a dead time of approximately $2.3 \mu\text{s}$.

The three values from the integrator are converted individually by a 12-bit Analogue to Digital Converter (ADC). These three values along with the number of clock cycles between the coincident pulses (determined by RUN logic signals) and two other counters form an event. One of the two “other” counters records the number of times the RUN threshold was crossed and the other counts the number of times a coincidence was observed. These two counters give information on the number of background events. The event is stored in a four-level FIFO, before being read out by the system, which is now described.

3.3 Read out of the system

Each board had a unique address (integer from 1 to 8) associated with the DAQ software, which also acted as an identifier in the data. The backplane of the crate supplied each board with 5 V (DC) and provided the read out to the Data Acquisition (DAQ) PC via a PCI card. The PCI card used in this system was a National Instruments PCI-6503 digital IO card which had been developed to run on Windows XP. Information from the boards was written by the DAQ system to a data file that recorded the address of the board that detected the coincidence, time since the start of the run, time interval between the pulses, integral of the beta pulse (ADC counts), integral of the alpha pulse (ADC counts), fraction of charge in the tail of the pulse and date. In addition to the data file, the DAQ stored a log file containing information about samples being counted by the system, PMT high voltage and any other necessary information.

3.4 System calibration

Before the system was used for the first time, a pedestal check was performed to measure the zero-offset of each discriminator. Once complete, the pedestal values for each board were stored in a file which was loaded by the DAQ system each time it was used. The pedestal check was repeated every few months and no significant variation in their values was found.

To define the signal and sources of background, 50 ml liquid scintillator sources were manufactured using 42 g of Optiphase HiSafe-3 liquid scintillator cocktail and an acidified solution of either ^{228}Th , ^{226}Ra or a mixture of both. The source strengths used to define the signal ranged from 0.5 mBq to 2 Bq, and a set of “blank” samples which contained 42 g of Optiphase HiSafe-3 and 10 mL of 0.5 mol/L hydrochloric acid were used to determine the background. Plotting the ADC value of the second pulse of the coincidence against the fraction of charge in the tail of the second pulse yields clear separation between signal and background.

A relatively strong ^{228}Th source was used to ensure that the alpha peak from ^{212}Po occurred between 900 and 1000 ADC counts and the tail fraction peak was at a value of around 0.20. The

³A 16 bit counter in the FPGA is used to determine this time.

rate of events was confirmed to be similar for all eight channels of the system. Once complete, a ^{226}Ra source was used to confirm that nearly all of the alpha energy (ADC) spectrum was seen for ^{210}Po and that the tail fraction peak also occurred close to 0.20 for ^{238}U chain coincidences.

Once the peak positions had been verified, a blank sample was used to measure the background count rate of the system. Each background run took approximately 1 week to acquire enough statistics to provide a good measurement. The background spanned the same ADC values as the signal, but had a much lower value of the tail fraction, with a maximum peak at around 0.125.

A plot of ADC value versus tail fraction for the second event in a coincidence showed clear separation between signal and background. Once the signal and background regions had been determined using the high rate source and blank sample, a low rate source (0.5 to 5.0 mBq) was counted. Such sources were usually a mixture of ^{226}Ra and ^{228}Th and were strong enough to show a clear signal, but weak enough to show the background. A plot of activity for the Th chain in a mixed Ra/Th source is shown in figure 3. These provided the best measurement of signal and background separation and were used to measure the counter efficiency.

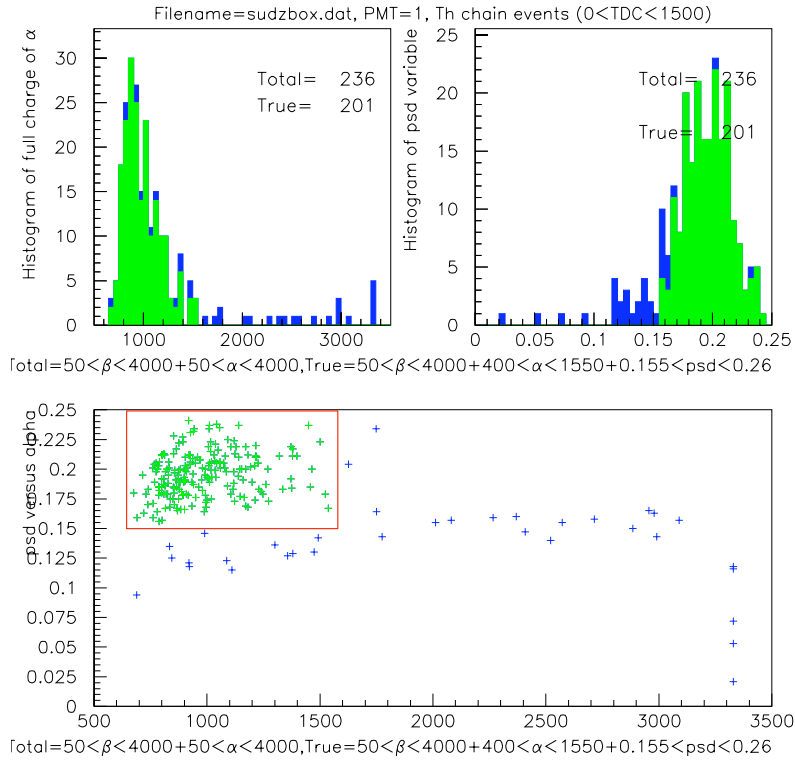


Figure 3. Thorium chain results from a typical run using a 5 mBq ^{228}Th source. On the lower plot, the signal are the points that are contained within the (red) rectangle in the upper left hand corner. Points outside the (red) box are background events. The maximum value of the alpha ADC was approximately 3320 counts.

To determine the counting efficiency, a known strength source was counted and the measured activity compared with the actual source strength. The counting efficiency, ϵ was defined as

$$\epsilon = \frac{R_m}{R_a}. \quad (3.1)$$

where R_m is the measured source strength and R_a the actual source strength.

Tail fraction and ADC cuts were applied to the data to select only genuine beta-alpha coincidences. The activity of the sample was determined by fitting time spectra to the rate of these selected events as a function of time since the start of counting. A maximum likelihood method was used and the contribution of all radioisotopes in the chain was taken into account. Using this method, the efficiency of the system was independent of channel number and measured to be:

$$\begin{aligned} 50 \pm 5\% & \text{ for } ^{224}\text{Ra} \\ 62 \pm 7\% & \text{ for } ^{226}\text{Ra} \end{aligned}$$

The branching ratio was included in the ^{224}Ra efficiency. As the cuts used to select coincidences were optimised for ^{224}Ra counting, the efficiency of ^{226}Ra detection is lower than expected. The energy of the ^{212}Po alpha is 8.95 MeV and that for ^{210}Po is 7.83 MeV. The lower bound on the alpha energy was optimised for the ^{212}Po alpha which resulted in some U chain alphas being cut. In addition, the longer time window required to observe the ^{214}Bi - ^{210}Po coincidences resulted in a higher background of random coincidences for U chain events. It was therefore necessary to use a more stringent PSD cut to reject backgrounds. This coupled with the fact that PSD decreases slightly as energy decreases makes the selection of U chain events more difficult and leads to a lower efficiency for ^{226}Ra events.

4 Assay results

Towards the end of the third phase of the SNO experiment, several assays were counted using this beta-alpha system. To compare the performance of this system to the previous NIM system, an assay of water in the cavity region of the detector was performed, the sample split and part counted on each system. Taking into account small differences in the masses of the samples, the equivalent concentrations of ^{232}Th were

$$\begin{aligned} 21.0_{-7.0}^{+7.5} \times 10^{-14} \text{ gTh/gH}_2\text{O} & \text{ for the new system} \\ 17.0_{-6.6}^{+6.9} \times 10^{-14} \text{ gTh/gH}_2\text{O} & \text{ for the old system.} \end{aligned}$$

As the sample was split between two systems, the statistical error was larger than usual. Despite this, good agreement between the two systems was found. When combined the results were in good agreement with previous measurements of water radioactivity in the cavity region.

A second cavity assay using the standard HTiO assay procedure [4] was performed just after the official end of the NCD data-taking period. The sample was counted using the new system and the activity was found to be

$$96.0_{-28}^{+31} \times 10^{-14} \text{ gTh/gH}_2\text{O}.$$

As this assay was performed just after the final shutdown of the SNO experiment, it is suspected that the removal of test equipment located near the assay sampling point may have disturbed some radioactivity which could have been extracted by the assay. This led to a higher central value compared with the split sample assay.

5 Conclusions

A portable beta-alpha counting system for use with small aqueous samples was developed at the University of Oxford for use in the SNO experiment. Following extensive testing and calibration of this system, the efficiency was measured to be $50 \pm 5\%$ for ^{224}Ra and $62 \pm 7\%$ for ^{226}Ra . Direct comparison between this new and the previous system demonstrated that their performances were comparable. The success of the assay and calibration source measurements have led to the use of this beta-alpha counting system in a number of low-background particle physics experiments, including SNO+, Daya Bay and Picasso. Following the end of the SNO experiment, the counting system was donated to the SNOLAB facility where it is an active part of their low-level counting program.

Acknowledgments

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